



# **Technical Language Service**

Translations From And Into Any Language

## **JAPANESE / ENGLISH TRANSLATION OF**

**Japanese Patent Application JP 1 – 138260 A**

**Title: Polyester Resin Composition and Method for  
Manufacturing Same**

**Your Ref: No. 11681**

**For: Eastman Chemical Company -  
Library and Information Services (LibrIS)**

## (12) Unexamined Patent Gazette (A)

1-138260

(51) Int. Cl. <sup>4</sup>	Classification Symbols	Internal Office Registration Nos.	(43) Date of Publication: May 31, 1989
C 08 L 67/00	LPA	A-7224-4J	
C 08 K 5/09	CAM		
5/17	CAM		
C 08 L 67/00	KJV	C-7224-4J	
	KJX	B-7224-4J	
Request for Examination: Not yet submitted		Number of Inventions: 2	Total of 8 pages [in original]

(54) Title of the Invention: **Polyester Resin Composition and Method for Manufacturing Same**

(21) Application No.: 62-296991  
(22) Date of Filing: November 24, 1987  
(72) Inventor: Toshiyuki Hirose (3-5 Misono 1-chome, Otake-shi, Hiroshima-ken)  
(72) Inventor: Shigenobu Otsubo (6-9 Misonodai, Otake-shi, Hiroshima-ken)  
(71) Applicant: Mitsui Petrochemical Industries, Ltd. (2-5 Kasumigaseki 3-chome, Chiyoda-ku, Tokyo)  
(74) Agent: Shunichiro Suzuki, Patent Attorney

### SPECIFICATION

#### 1. Title of the Invention

Polyester Resin Composition and Method for Manufacturing Same

#### 2. Claims

(1) A polyester resin composition obtained as a result of mixing the following components in the relative amounts indicated:

- (i) 100 weight parts of thermoplastic polyester resin,
- (ii) 1 to 100 weight parts of a polar group-containing polyolefin,
- (iii) 0.01 to 10 weight parts of an amine compound, and
- (iv) 0.01 to 20 weight parts of a metal salt of an organic acid.

- (2) A method for manufacturing a polyester resin composition, characterized in that
- (i) 100 weight parts of thermoplastic polyester resin are melted and mixed with
  - (ii) a polar group-containing polyolefin added thereto in an amount of 1 to 100 weight parts, followed by the addition, melting, and kneading of
  - (iii) 0.01 to 10 weight parts of an amine compound and
  - (iv) 0.01 to 20 weight parts of a metal salt of an organic acid.

### **3. Detailed Description of the Invention**

#### **Technological Field of the Invention**

The present invention relates to a thermoplastic polyester resin composition capable of yielding molded articles that have adequate moldability, excellent appearance, and superior impact resistance and other mechanical properties, and to a method for manufacturing such a composition.

#### **Technological Background of the Invention, and Problems Thereof**

Polyester resins have excellent mechanical properties, electrical properties, heat resistance, chemical resistance, and the like, and are used in a variety of commercial products. However, such resins have low impact resistance, and notched impact strength in particular, and possess inferior moldability, so numerous improvement techniques have conventionally been proposed. For example, JP (Kokai) 60-40154 discloses a method in which an ethylene/ $\alpha$ -olefin copolymer grafted with an epoxy monomer is blended with polyester resin; JP (Kokai) 58-17148 discloses a method in which an ethylene/ $\alpha$ -olefin copolymer and a glycidyl-containing copolymer composed of an  $\alpha$ -olefin and a glycidyl ester of an  $\alpha,\beta$ -unsaturated acid are blended with a polyester resin; and JP (Kokai) 58-38747 discloses a method in which an alicyclic carboxylic acid-modified olefinic elastomer is blended with a polyester resin. The polyester resin compositions thus obtained possess significantly improved impact resistance.

However, the polyester resin compositions disclosed in such publications still have inadequate moldability. Specifically, polyethylene terephthalate and other polyester resins have a low polymerization rate, so the degrees of polymerization are different on the surface and in

the interior of a molded article, and its appearance, mechanical properties, dimensional stability, and shape stability are compromised. In addition, it is impossible to obtain products with adequate heat resistance because of insufficiently advanced crystallization. In view of this, a polyester resin composition obtained as a result of blending in a nucleator and a plasticizer in order to improve the moldability of polyethylene terephthalate has been proposed. For example, JP (Kokai) 58-217547 discloses a polyester resin composition obtained as a result of blending a sodium or potassium salt of a copolymer of an olefin and acrylic acid or methacrylic acid as a nucleator together with polyethylene terephthalate and an inorganic filler, and then further blending an ester of an alcohol and an aliphatic carboxylic acid as a plasticizer. This polyester resin composition does indeed have excellent moldability but the resin surface is rough because of the presence of inorganic filler, and the resin is unsuitable for applications in which an improved appearance is required. In addition, adding a nucleator and a plasticizer to a composition comprising a polyester and a modified olefinic elastomer in the manner disclosed in JP (Kokai) 58-217547 yields a composition with low impact strength.

The inventors have discovered that a method in which a polyester resin and known polyester resin modifiers are kneaded together in order to overcome the drawbacks of such polyester resins is an effective technique, and have disclosed this technique in Japanese Patent Application (Tokugan) 61-274154. Developing a polyester resin that would have even better impact strength was still required for some applications, however.

### Object of the Invention

The present invention is designed to solve the drawbacks associated with prior art as described above, and an object thereof is to provide a thermoplastic polyester resin composition that endows molded articles with adequate surface characteristics, has excellent impact resistance and other mechanical properties, possesses excellent thermal resistance and other characteristics, and exhibits adequate moldability.

### Summary of the Invention

The polyester resin composition according to the present invention is obtained as a result of mixing the following components in the relative amounts indicated:

- (i) 100 weight parts of thermoplastic polyester resin,
- (ii) 1 to 100 weight parts of a polar group-containing polyolefin,
- (iii) 0.01 to 10 weight parts of an amine compound, and
- (iv) 0.01 to 20 weight parts of a metal salt of an organic acid.

The method for manufacturing a polyester resin composition according to the present invention is characterized in that

- (i) 100 weight parts of thermoplastic polyester resin are melted and mixed with
- (ii) a polar group-containing polyolefin added thereto in an amount of 1 to 100 weight parts, followed by the addition, melting, and kneading of
- (iii) 0.01 to 10 weight parts of an amine compound and
- (iv) 0.01 to 20 weight parts of a metal salt of an organic acid.

In the present invention, an amine compound and a metal salt of an organic acid are added to a polyester resin composition according to a specified kneading method, yielding a thermoplastic polyolefin resin composition that has adequate surface characteristics and improved impact resistance, thermal resistance, and moldability.

#### Detailed Description of the Invention

The components of the thermoplastic polyester resin composition and the manufacturing method of the present invention are described in detail below.

#### Thermoplastic Polyester Resin

The thermoplastic polyester resin (i) used in the present invention is mainly polyethylene terephthalate, but a portion of the terephthalic acid component or the diol component may be substituted with another copolymerization component. Examples of the copolymerization component that may be used include isophthalic acid, naphthalene dicarboxylic acid, 4,4-diphenoxyethane dicarboxylic acid, adipic acid, sebacic acid, cyclohexanedicarboxylic acid, and other bifunctional dicarboxylic acids. Examples of the diol component that may be used include ethylene glycol, trimethylene glycol, tetramethylene glycol, hexamethylene glycol, polyethylene glycol, polypropylene glycol, polytetramethylene glycol, copolymers of polyethylene glycol and polypropylene glycol, and the like. The polyester resin (i) used in the

present invention may be a mixture of two or more types of polyester resins such as those described above.

A preferred polyester is a crystalline thermoplastic polyester having at least 80 mol% ethylene terephthalate repeating units or polyethylene terephthalate.

(iv)<sup>1</sup> Polar Group-containing Polyolefin

The polar group-containing polyolefin used in the present invention is a polyolefin having at least one polar group selected from carboxyl groups, carboxylic acid anhydride groups, and epoxy groups. Such a polar group-containing polyolefin can be manufactured by means of copolymerizing an olefin monomer with a vinyl monomer having a carboxyl group or an anhydride thereof in the molecule, or with a vinyl monomer having an epoxy group in the molecule; as a result of graft polymerizing a vinyl monomer such as that described above onto an olefin polymer; as a result of oxidizing a double bond in polybutadiene, polyisoprene, or another polydiene by means of a known method to introduce an epoxy group; or with the help of another method.

Examples of the carboxyl group-containing vinyl monomer used when introducing a carboxyl group into the polyolefin include acrylic acid, methacrylic acid, maleic acid, fumaric acid, itaconic acid, citraconic acid, tetrahydrophthalic acid, bicyclo [2,2,1] hepto-2-n-5,6-dicarboxylic acid, and other unsaturated carboxylic acids, as well as maleic anhydride, itaconic anhydride, citraconic anhydride, tetrahydrophthalic anhydride, bicyclo [2,2,1] hepto-2-n-5,6-dicarboxylic acid anhydride, and other anhydrides of unsaturated carboxylic acids. Particularly preferred are maleic anhydride and fumaric acid.

Maleic anhydride, itaconic anhydride, citraconic anhydride, tetrahydrophthalic anhydride, bicyclo [2,2,1] hepto-2-n-5,6-dicarboxylic acid anhydride, and other anhydrides as described above are used to introduce a carboxyl group into the polyolefin.

Examples of the vinyl monomer that has an epoxy group in the molecule and is used when introducing an epoxy group into the polyolefin include glycidyl acrylate, glycidyl

---

<sup>1</sup> Should probably read (ii)

methacrylate, monoglycidyl itaconate, butene dicarboxylic acid diglycidyl ester, butene dicarboxylic acid monoglycidyl ester, 2-methylacryl glycidyl ether, vinyl-glycidyl ether, 3,4-epoxy-3-methyl-1-butene, vinyl cyclohexene monoxide, and p-glycidyl styrene. Of these, glycidyl methacrylate is preferred.

Examples of the olefin polymer that serves as the base for the polar group-containing polyolefin include noncrystalline, low crystallinity, or crystalline olefin polymers in which ethylene, propylene, 1-butene, 1-pentene, 4-methyl-1-pentene, 1-hexene, 1-octene, 1-decene, 1-dodecene, 1-tetradecene, 1-hexadecene, 1-octadecene, 1-eicosen, or another  $\alpha$ -olefin component is the main component. Additional examples include polymers in which small quantities of the following components are randomly polymerized, graft polymerized, or added besides the  $\alpha$ -olefin components: butadiene, isoprene, 1,4-hexadiene, 5-ethylidene-2-norbornene, 5-vinyl-2-norbornene, or another diene component; vinyl acetate, acrylic acid (salt), methacrylic acid (salt), glycidyl acrylate, ethyl acrylate, or another acrylic acid ester; glycidyl methacrylate, ethyl methacrylate, or another methacrylic acid ester; maleic acid, maleic anhydride, maleic acid ester, 2-norbornene-5,6-dicarboxylic acid, 2-norbornene-5,6-dicarboxylic acid anhydride, or another unsaturated carboxylic acid; derivative components thereof such as acrylonitrile, methacrylonitrile, and other nitrile components; and chlorine, bromine, iodine, and other halogens.

Homopolymers, copolymers, or hydrogenation products in which butadiene, isoprene, or another diene is the main component may also be used as the olefin polymer.

Among these olefin polymers, specific examples of noncrystalline or low-crystallinity olefin polymers include copolymers of ethylene and propylene; copolymers of ethylene and 1-butene; copolymers of ethylene and 1-hexane; copolymers of propylene and 1-butene; copolymers of propylene, 1-butene, and 4-methyl-1-pentene; copolymers of 1-hexene and 4-methyl-1-pentene; copolymers of ethylene, propylene, and dicyclopentadiene; copolymers of ethylene, propylene, and 5-ethylidene-2-norbornene; copolymers of ethylene, propylene, and 5-vinyl-2-norbornene; copolymers of ethylene, propylene, and 1,4-hexadiene; copolymers of ethylene, 1-butene, and dicyclopentadiene; copolymers of ethylene, 1-butene, and

5-ethylidene-2-norbornene; copolymers of ethylene, 1-butene, and 1,4-hexadiene; polybutadiene and hydrogenation products thereof; and polyisoprene and hydrogenation products thereof.

Specific examples of the crystalline olefin polymer include polyethylene, polypropylene, poly-1-butene, poly-4-methyl-1-pentene, copolymers of ethylene and propylene, copolymers of ethylene and 1-butene, copolymers of ethylene and 4-methyl-1-pentene, copolymers of ethylene and 1-hexene, copolymers of ethylene of vinyl acetate, copolymers of ethylene and acrylic acid (salt), copolymers of ethylene and methacrylic acid (salt), copolymers of ethylene and glycidyl acrylate, copolymers of ethylene and glycidyl methacrylate, copolymers of ethylene and maleic acid, and copolymers of ethylene and maleic anhydride.

Additional examples of olefin polymers include olefin polymers in which acrylic acid, methacrylic acid, methacrylic acid ester, maleic acid, maleic anhydride, 2-norbornene-5,6-dicarboxylic acid, 2-norbornene-5,6-dicarboxylic acid anhydride, and other unsaturated carboxylic acids or derivative components thereof are graft polymerized to the olefin polymers cited above.

The content of the polar group in such a polar group-containing polyolefin varies considerably in accordance with the type of vinyl monomer that is used, but 0.01 to 20 wt% is commonly preferred.

A polar group content of less than 0.01 wt% is not preferred because the improved impact resistance derived from blending a polar group-containing olefin is inadequate, and a content exceeding 20 wt% is not preferred because the impact strength tends to decrease after the addition of a nucleating agent.

The intrinsic viscosity  $[\eta]$  of these olefin polymers, as measured in decalin at a temperature of 135°C, is 0.05 to 30 dL/g, and is preferably 0.1 to 25 dL/g.

Also possible in the present invention is the use of a mixture of polar group-containing polyolefins such as those described above and polyolefins that do not contain a polar group.



### Amine Compounds (iii)

The amine compound used in the present invention is a compound having one or more primary amine group or secondary amine group per molecule. Specific examples of such amine compounds include propylamine, isopropylamine, butylamine, hexylamine, cyclohexylamine, and other aliphatic primary amines; aniline, toluidine, xyldine, and other aromatic primary amines; dimethylamine, diethylamine, dipropylamine, diisopropylamine, dibutylamine, dipentylamine, dicyclohexylamine, piperidine, n-methylaniline, ethylenediamine, hexamethylenediamine, diethylenetriamine, triethylenetetramine, and other polyhydric amines; and ethanolamine, propanolamine, and other alkanolamines. Among these, aliphatic primary amines and aliphatic secondary amines are preferred because they do not compromise the moldability of the polyester resin composition.

Blending these types of amine compounds with the polyester resin composition allows the impact strength of the resin composition to be further enhanced.

### Metal Salt of Organic Acid

The metal salt of an organic acid used in the present invention is added for the purpose of promoting the crystallization velocity of the polyester. In other words, the organic acid metal salt may be thought of as contributing to the formation of polyester crystal nuclei.

Examples of organic acids that can be used in the formation of an organic acid metal salt include formic acid, acetic acid, stearic acid, montanic acid, oleic acid, linoleic acid, oxalic acid, phthalic acid, isophthalic acid, terephthalic acid, tartaric acid, salicylic acid, benzoic acid, and carbonic acid. Preferable among these is benzoic acid. Examples of metals that can form the salts include sodium, potassium, calcium, magnesium, barium, and zinc.

Sodium, potassium, calcium, zinc, and other salts of copolymers of olefins or styrene and acrylic acid or methacrylic acid may be used as the organic acid metal salt. Of these two, the latter is preferred. Organic acid metal salts of these may be used alone or as a mixture of two or more types.

### Blending Ratio of the Components

In the polyester resin composition related to the present invention, components such as those described above are included in ratios such as those indicated below with respect to 100 parts by weight of thermoplastic polyester resin (i).

#### Polar Group-containing Polyolefin (ii)

The polar group-containing polyolefin is contained in an amount of 1 to 100 parts by weight, and preferably 3 to 90 parts by weight, with respect to 100 parts by weight of the thermoplastic polyester resin. When the amount of polar group-containing polyolefin is less than 1 part by weight with respect to 100 parts by weight of the thermoplastic polyester resin, the impact resistance of the resulting polyester resin composition is improved only slightly, so this ratio is not preferred; and when the amount of polar group-containing polyolefin exceeds 100 parts by weight, the mechanical properties of the resulting polyester resin composition are adversely affected, so this ratio is not preferred either.

#### Amine Compound (iii)

The amine compound is contained in an amount of 0.01 to 10 parts by weight, preferably 0.01 to 7 parts by weight, and more preferably 0.05 to 5 parts by weight, with respect to 100 parts by weight of the thermoplastic polyester resin. When the amount of amine compound is less than 0.01 parts by weight with respect to 100 parts by weight of the thermoplastic polyester resin, the impact resistance of the resulting polyester resin composition is improved only slightly, so this ratio is not preferred; and when the amount of amine compound exceeds 10 parts by weight, the mechanical properties of the resulting polyester resin composition are adversely affected, so this ratio is not preferred either.

#### Metal Salt of Organic Acid (iv)

The metal salt of an organic acid is contained in an amount of 0.01 to 20 parts by weight, and preferably 0.1 to 10 parts by weight, with respect to 100 parts by weight of the thermoplastic polyester resin. When the amount of metal salt of an organic acid is less than 0.01 parts by weight with respect to 100 parts by weight of the thermoplastic polyester resin, the effect of

promoting the crystallization velocity of the polyester tends to be reduced, so this ratio is not preferred; and when the amount of metal salt of an organic acid exceeds 20 parts by weight, the mechanical properties or the thermal resistance properties of the resulting thermoplastic polyester resin composition are adversely affected, so this ratio is not preferred either.

#### Other Components

In addition to the above-described components, the following may be added to the polyester composition of the present invention: organic or inorganic compounds such as plasticizers, antioxidants, UV absorbers, other stabilizers, lubricants, flame retardants, antistatic agents, coloring agents, mold release agents, glass fiber, talc, and other fillers.

The plasticizer is preferably an ester of an aliphatic alcohol and a carboxylic acid. Specific examples of such plasticizers include adipic acid-di-(2-ethylhexyl) ester, adipic acid-di-n-butyl ester, poly (butane 1,3-diol adipate), poly (hexane 1,6-diol adipate), poly (butane 1,4-diol adipate), montanic acid ester, alkylsulfonic acid-p-phenyl esters, diethylene glycol-di-benzoate, neopentyl glycol-di-benzoate, pentaerythritol dibenzoate, and thio-di-ethanol-di-benzoate. Such plasticizers may be used alone or as a mixture of two or more types. The blending ratio of such plasticizers is ordinarily 0.01 to 20 parts by weight, and more preferably 0.1 to 10 parts by weight, with respect to 100 parts by weight of the polyester resin.

Polyolefins that do not contain a polar group may be added to the polyester composition of the present invention.

#### Manufacturing Method

The polyester resin composition of the present invention can be manufactured as a result of melting and kneading a thermoplastic polyester resin (i) and a polar group-containing polyolefin (ii) such as those described above, thereafter adding an amine compound (iii) and a metal salt of an organic acid (iv), and then melting and kneading the product. The order in which the amine compound (iii) and metal salt of an organic acid (iv) are added is not particularly limited, but the amine compound (iii) is preferably added to the melted and kneaded product of

the thermoplastic polyester resin (i) and the polar group-containing polyolefin (ii) prior to adding the metal salt of an organic acid (iv).

The impact resistance of polyester resin obtained as a result of kneading the above-described four components (i), (ii), (iii), and (iv) cannot be improved with the help of methods other than those described above.

Following are the possible reasons that the thermoplastic polyester resin (i) and polar group-containing polyolefin (ii) must first be melted and kneaded, and an amine compound (iii) and a metal salt of an organic acid (iv) must then be added as needed according to the present invention.

In accordance with a study conducted by the inventors, the above-described four components react with each other in the melting and kneading steps, and the reaction between the thermoplastic polyester resin (i) and the polar-group containing polyolefin (ii) enhances the dispersibility of (ii) in (i) and improves the impact resistance of the composition. It was found, however, that the reaction between the polar-group containing polyolefin (ii) and the metal salt of an organic acid (iv) forms crosslinks in the polar-group containing polyolefin (ii) and markedly hinders the dispersibility of (ii) in (i). In other words, in order to knead the four components and obtain a composition with excellent impact resistance, (i) and (ii) must be melted and kneaded in advance to cause a reaction, and the reaction between (ii) and (iv) must be prevented by means of adding (iv) thereafter. It was found that a small amount of reaction residue may be present even when (i) and (ii) have been kneaded in advance, and the reaction between (iv) and this reaction residue may also inhibit the dispersibility of (ii).

As a result of studying additives for rendering this reaction residue inactive, an amine compound (iii) was found to be effective. In other words, it is thought that the amine compound (iii) reacts with the reaction residue (ii) and renders (ii) inactive.

The kneading in the method for manufacturing the composition of the present invention may be carried out with known means. The kneading may be carried out in an extruder, kneader, or Banbury mixer, for example.

It is preferable from an aspect of practicality to load the thermoplastic polyester resin (i) and the polar-group containing polyolefin (ii) into an extruder hopper and to feed (iii) and (iv) from an intermediate position of the extruder cylinder.

### Effect of the Invention

The polyester composition obtained in accordance with the present invention can easily be formed as a result of injection molding, extrusion molding, or another ordinary method. In particular, molded articles with adequate appearance and excellent mechanical properties can be obtained even if the mold is used at a temperature of 130°C or less during injection molding. The polyester composition of the present invention may be used in a variety of configurations and applications, but its use is particularly advantageous in materials for exterior automotive panels and other fields that require impact resistance, high rigidity, and high thermal resistance.

The present invention is described below with working examples, but the present invention is not limited by the working examples.

### Working Examples 1 to 6

#### [I] Method of Manufacturing the Composition

Polyethylene terephthalate with an intrinsic viscosity of 0.65 dL/g, a polar group-containing polyolefin manufactured in accordance with the reference example described below, and optional EBR were mixed in the ratio shown in Table 1, fed to a twin-screw extruder set to a temperature of 260°C, and then formed into pellets. The pellets were dried in a reduced-pressure dryer, n-hexylamine was added to the pellets, the pellets were then kneaded with an extruder, a sodium salt (ethylene/methacrylic acid weight ratio: 85/15) of an ethylene/methacrylic acid copolymer with a neutralization degree of 60% (nucleating agent) and diethylene glycol dibenzoate (plasticizer) were added to the resulting pellets, the product was again extruded at 260°C from the extruder, and the extrudate was injection-molded at a mold temperature of 120°C (sequential blending method).

Shown in comparative example 1 of Table 1 are the results for polyethylene terephthalate in which the components of Table 1 were dry blended and extruded all at once (batch blending method).

### [III] Method of Manufacturing the Polar-Group Containing Polyolefin

#### Reference Example 1

100 parts by weight of an ethylene-butene copolymer that had an ethylene content of 85 mol% and a melt index of 3.60 g/10 min at 190°C, and 1 part by weight maleic anhydride were mixed with 0.06 parts by weight of 2,5-dimethyl-2,5-di(tertiary butyl peroxy)hexyne-3, and the product was fed to a single-screw extruder (diameter: 25 mm; L/D:28) and extruded at a temperature of 250°C to obtain a maleic anhydride-modified ethylene-butene copolymer (hereinafter abbreviated as "EBR"). The modified EBR was dissolved in xylene, reprecipitated with acetone, and then dried. The amount in which the maleic anhydride had been grafted was measured with the titration method and found to be 0.45 wt%.

#### Reference Example 2

Other than replacing the maleic anhydride used in reference example 1 with glycidyl methacrylate, glycidyl methacrylate-grafted EBR was obtained with the help of the same procedure as reference example 1. The graft amount of glycidyl methacrylate was 0.50 wt%.

#### Reference Example 3

Bondfast-E<sup>®</sup> manufactured by Sumitomo Chemical was used as the ethylene-glycidyl methacrylate copolymer.

### [II] Method of Evaluating the Composition

MFR was measured in accordance with ASTM D-1238. The flexural elastic modulus (FM) and flexural strength (FS) were measured in accordance with ASTM D-790-80 using a sample with a thickness of 1/8". Izod impact strength (IZ) was measured in accordance with ASTM D-256 using a notched sample with a thickness of 1/8". The heat deformation temperature (HDT) was measured in accordance with ASTM D-256 under a load of 66 PSI. The

crystallization velocity ( $\Delta H_H/\Delta H_C$ ) was measured using a differential thermal analyzer (DSC). A sample was placed in a mold with a thickness of 1 mm, pressed at a temperature of 290°C under a load of 60 kg/cm<sup>2</sup>, and thereafter cooled to obtain a pressed sheet. A DSC measurement was carried out under conditions in which a sample of about 10 mg was taken from the center portion of the pressed sheet, heated at a rate of 10°C/min, held for 10 minutes at 290°C, and cooled at a rate of 10°C/min. The amount of heat  $\Delta H_H$  calculated from the peak surface area at the crystallization temperature when the temperature was increasing, and the amount of heat  $\Delta H_C$  calculated from the peak surface area at the crystallization temperature when the temperature was decreasing were measured from the resulting thermal curve. The thus obtained  $\Delta H_H/\Delta H_C$  is an index of the ease of crystallization. The smaller this value is, the greater the ease with which the polyester crystallizes.

Table 1

		Working Example 1	Working Example 2	Working Example 3	Working Example 4
Blend	Polyethylene terephthalate (parts by weight)	80	80	80	80
	Polar group-containing polyolefin	Type	Polymer cited in reference example 1	Polymer cited in reference example 2	Polymer cited in reference example 3
		Polar group content (wt%)	0.45	0.45	0.50
		MFR <sup>190°C</sup> (g/min)	1.0	1.0	1.0
		Blend amount (parts by weight)	20	20	20
	EBR (parts by weight)	0	0	0	10
	n-Hexylamine (parts by weight)	0.1	0.2	0.2	0.2
	Nucleating agent (parts by weight)	3	3	3	3
	Plasticizer (parts by weight)	4	4	4	4
	Blending method	Sequential blending	Sequential blending	Sequential blending	Sequential blending
	MFR <sup>260°C</sup> (g/10 min)	18	19	18	15
	FM (kg/cm <sup>2</sup> )	15,700	15,800	15,800	16,000
	FS (kg/cm <sup>2</sup> )	500	510	530	520
	Notched IZ <sup>23°C</sup> FM (kgcm/cm)	38	42	40	50
	HDT (°C)	145	143	144	140
	$\Delta H_H/\Delta H_C$	0.21	0.20	0.22	0.22
	Appearance	○	○	○	○

Table 1 (continued)

		Working Example 5	Working Example 6	Working Example 7	Working Example 8
Blend	Polyethylene terephthalate (parts by weight)		80	80	80
	Polar group- containing polyolefin	Type	Polymer cited in reference example 1	Polymer cited in reference example 1	Polymer cited in reference example 2
		Polar group content (wt%)	0.45	0.45	0.45
		MFR <sup>190°C</sup> (g/min)	1.0	1.0	1.0
		Blend amount (parts by weight)	20	20	20
	EBR (parts by weight)		0	0	0
	n-Hexylamine (parts by weight)		0.2	0.2	0.2
	Nucleating agent (parts by weight)		3	3	0
	Plasticizer (parts by weight)		0	4	4
	Blending method		Sequential blending	Sequential blending	Sequential blending
	MFR <sup>260°C</sup> (g/10 min)		17	15	18
	FM (kg/cm <sup>2</sup> )		15,600	16,000	16,000
	FS (kg/cm <sup>2</sup> )		500	550	530
	Notched IZ <sup>23°C</sup> FM (kgcm/cm)		37	18	2.8
	HDT (°C)		144	141	104
	$\Delta H_H/\Delta H_C$		0.30	0.25	0.53
	Appearance		○	○	×